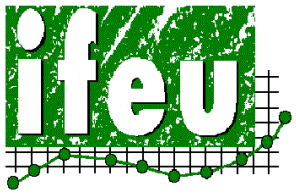

Review of Radiological Monitoring at LBNL

Preliminary Technical Report

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Prepared under Contract with the City of Berkeley

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Executive Summary

This preliminary technical report describes work in progress performed by the Institute for Energy and Environmental Research based in Heidelberg, Germany (IFEU) under contract with the City of Berkeley, CA. The work addresses concerns regarding past and present radiation exposures from E.O. Lawrence Berkeley National Laboratory (LBNL). IFEU is reviewing data on present and past emissions and environmental monitoring by LBNL of soils, air, subsurface and ground waters, and plants (including raw data) in order to arrive at conclusions about both the quality of the data collected and the analyses of that data. Seventeen areas of concerns were identified and were divided into four groups: (a) exposures from current operations, (b) legacy contamination from past operations/Superfund issues, (c) historical exposures, and (d) risk related questions. Preliminary results are available for eight areas of concern. The results at this point can be summarized as follows:

1. Ambient air monitoring for tritium should be expanded. The number of monitoring sites at LBNL is well below the DOE average. At other DOE facilities with similar amounts of tritium emissions at least 16 wind directions (of 22.5 degrees) are monitored. A comparable network would be advisable for LBNL as well.
2. Releases of tritium are often in short bursts (e.g. 0.2 Ci of tritium emitted over 17 minutes on March 28, 1998). This renders the results of the computer program used to determine compliance with the NESHAP standard (CAP88PC) to be inaccurate. The probability that a person near the fence could receive a radiation dose of greater than 10 mrem/yr should be determined using appropriate models, accounting for the complex terrain and the discontinuous nature of the releases. However, there is no evidence at this time to suggest that offsite exposures resulted in radiation doses exceeding the 10 mrem/yr limit for any individual.
3. There are minor uncertainties associated with the measurements of tritium in ambient air at a given location. Some corrections are necessary.
4. Tritium inventory data is uncertain in the order of +/- 30% and not suitable to verify airborne releases. However, improved inventory data is useful to verify the type of operations at NTLF.
5. Historical data for tritium in ambient air is somewhat puzzling. Concentrations in ambient air do not correlate with reported releases, maximum levels were reported for Building 3 (Calvin) in 1977/1978. The integrated concentrations of tritium are similar to those at NTLF indicating the possibility that contamination of soil and groundwater may have occurred there as well. A preliminary sampling effort around the building is recommended.
6. With regard to historical radiation exposures, gamma and neutron doses are of the greatest concern. Levels at Olympus Gate may have exceeded then-prevailing dose limits in 1959 and 1960. Dose reconstruction was conducted at other DOE facilities where exposures to offsite residents were lower than the levels recorded for LBNL.
7. Based on a review of the tritium sampling plan, the following additions are suggested: (a) the ambient air monitoring network should be expanded of to cover all 16 wind direction sectors (of 22.5° each), (b) the HASL-300 core method should be used for soil sampling, whereby samples would be analyzed for additional depth increments, (c) groundwater sampling would be added in coordination with the State of California Water Resources Board.
8. EPA should provide a parallel calculation for the hazard ranking system if it is assumed that a larger population (such as the full-time equivalent visitor population) is entered for the location of the Lawrence Hall of Science.

The authors invite comments, which will be incorporated into the final report.

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1 Introduction

The E.O. Lawrence Berkeley National Laboratory (LBNL) is a multipurpose research facility located in the Berkeley/Oakland hills in Alameda County, California. It is operated by the University of California (UC) under contract with the U.S. Department of Energy (DOE). The site map of the 200 acre facility is shown in Figure 1.

In response to community concerns regarding radiation exposures from LBNL, the City of Berkeley contracted with the Institute for Energy and Environmental Research (IFEU)[1], based in Heidelberg, Germany to review radiological monitoring at Lawrence Berkeley National Laboratory. The concerns cover a wide range of issues, including past and present operations of the laboratory. The scope of work spelled out in the contract is attached in Appendix A.

The review process started with interviews of LBNL scientists and technicians, subcontractors, regulators, city officials, and representatives of the community to generate a list of concerns for evaluation. This took place during the week of February 28, 2000.

The authors also participated in the meetings of the Environmental Sampling Project Task Force meetings on March 1, April 25 and June 1, 2000, either in person or via teleconferencing. There were many communications via e-mail, telephone and mail between IFEU and the City of Berkeley, community representatives, LBNL personnel and contractors, as well as with staff of EPA's San Francisco Office. The authors wish to acknowledge the substantial documented input received from the Committee to Minimize Toxic Waste (CMTW) which helped focus the review. We also appreciate the willingness of LBNL to respond to many information requests through its contractor Dr. Owen Hoffman (SENES Oak Ridge, Inc.).

In the process, the authors obtained about 20,000 pages of documents for review. In addition, raw data relating to real-time monitoring of tritium releases from the NTLF stack was received. It is in the nature of a review process that additional questions always arise. Consequently, more material is expected for consideration over the next few months. The present report is a progress report. It presents a partial analysis of data evaluated thus far as a basis for further discussion. The work can by no means to be considered complete. Thus it is hoped that it may assist in clarifying some issues, and to prioritize areas of concern. The starting point is a list of concerns, which is presented in question format. The report attempts to consider all questions and outlines where work still needs to be done. It is in the nature of endeavors like this, that each question answered could give rise to a new one.

The reader should be aware of the fact that although the review attempts to cover a large array of questions, the finite resources of IFEU's consulting contract do not allow addressing every question in appropriate depth. The authors will appreciate any feedback that will be incorporated into the final report.

[1] The official legal German name of the Institute is "ifeu-Institut für Energie- und Umweltforschung Heidelberg GmbH". It is not part of the Institute for Energy and Environmental Research (IEER) in Takoma Park, MD where the principal author of this report was Executive Director from 1987 to 1998. Founded in 1988, IFEU is a non-profit research organization and has currently a staff of 30 scientists. More details about research areas and activities can be found on the web site (www.ifeu.de).

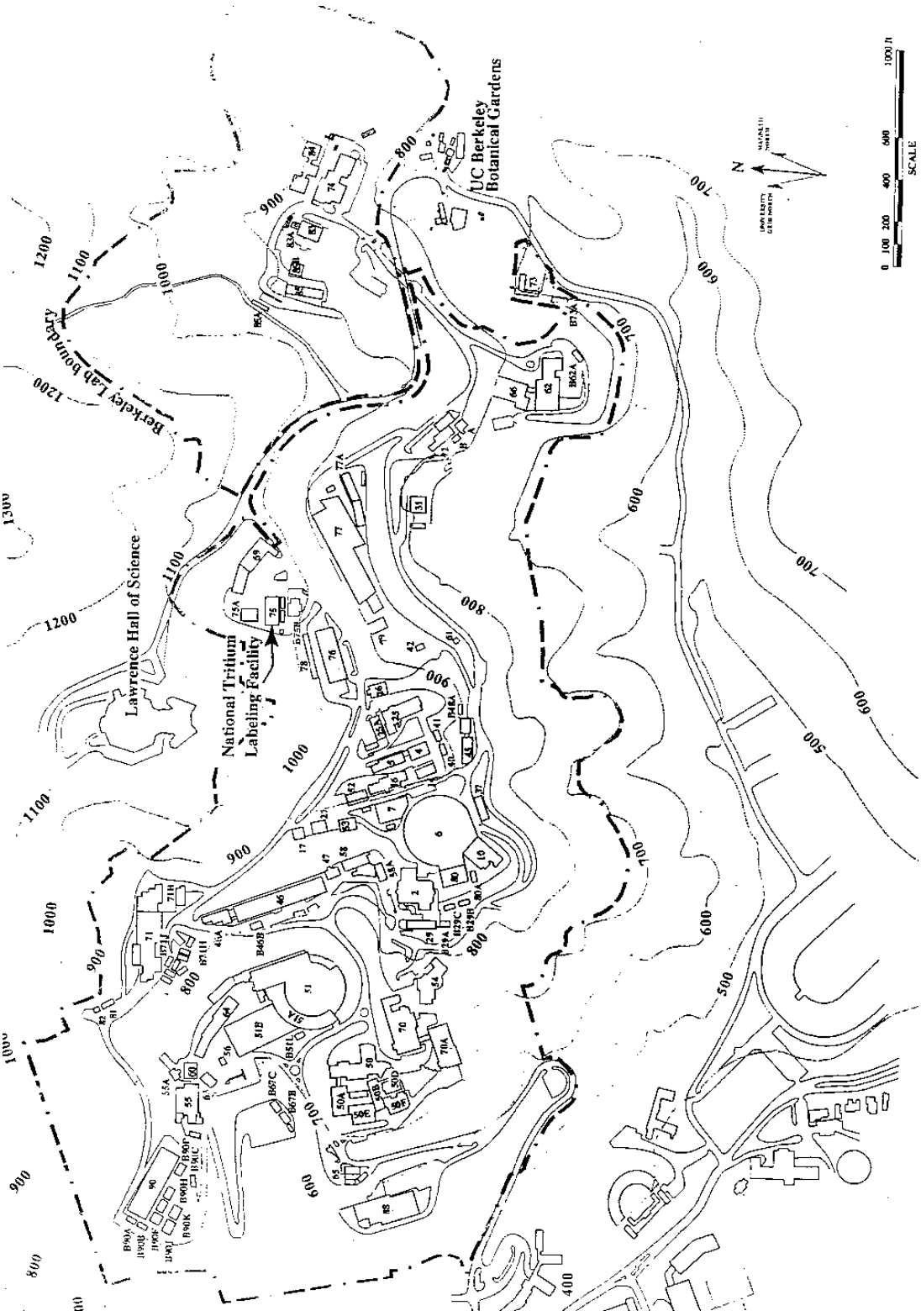

Figure 1. Site map of Lawrence Berkeley National Laboratory (from LBNL/DOE, 1999)

2 Identification of areas of concern

The major concerns of citizens regarding radiation exposures from LBNL operations which were broadly divided into four main categories for the purposes of this review:

A Concerns about current operations
B Concerns about legacy contamination from past operations
C Concerns about historical exposures
D Risk-related concerns

The authors attempted to approach the concerns as completely as possible. The presentation in question format is thought to allow the non-technical reader following the review process. Each chapter is organized in the following format:

- Concern
- Approach
- Findings to date
- Conclusions and recommendations

Table 1 and 2 contains a preliminary list of concerns. While some deal with rather specific technical issues, others cover larger problem areas. The list will be appended or revised as necessary.

Many questions currently focus on the activities at the National Tritium Labeling Facility (NTLF), in particular in connection with the Superfund evaluation by the US Environmental Protection Agency. In response to these activities, this report contains a section dealing with the LBNL/DOE Tritium Sampling and Analysis Plan, dated May 1999 (concern B.1 and B.2).

The evaluation of current operations is based on data for releases and environmental concentrations in the years 1998 and 1999. Data for earlier years are considered historical due to major changes in sampling and analytical techniques, quality control and documentation.

Table 1. Preliminary list of concerns relative to radiation exposures from current operations at LBNL

Concern	Approach	Status
A. Exposures from Current Operations		
A.1 Is the tritium inventory at NTLF adequately determined?	Review inventory data, its accuracy, and relevance to determine the environmental impacts of NTLF	Preliminary results
A.2 Are the releases of airborne tritium adequately monitored?	Review data on stack releases; evaluate internal consistency and uncertainties	Preliminary results
A.3 It tritium in air measured at the right locations?	Compare potentially affected locations with locations actually sampled	Preliminary results
A.4 Is the sampling and analysis of tritium in air at a given location sufficiently accurate?	Review observed versus expected water collected in silica gel samples Review results of split sampling program Review of contract laboratory performance	Preliminary results
A.5 Are radiation exposures to individuals (including sensitive subgroups) from NTLF operations below 10 mrem/yr including visitors to the area?	Review NESHAP compliance assessment; determine exposure scenarios that are not covered	In progress
A.6 Are model predictions used in NESHAP compliance assessment for the location of Lawrence Hall of Science accurate?	Review CAP88PC model input and output data, model limitations, and results of other models (CALPUFF)	In progress
A.7 Are measurements of discharges of radionuclides other than tritium into air and water from LBNL and the resulting radiation exposures sufficiently accurate?	Review of environmental monitoring data regarding radionuclides other than tritium	In progress
A.8 Are measurements of gamma and neutron radiation from LBNL sufficiently accurate?	Review environmental monitoring of gamma and neutron radiation	In progress

Table 2. Preliminary list of concerns relative to legacy contamination from past operations / Superfund Issues, historical exposures and risk related questions

Concern	Approach	Status
B. Legacy Contamination from Past Operations / Superfund Issues		
B.1 Is LBNL's Draft Tritium Sampling and Analysis Plan sufficient to determine the extent and nature of contamination at NTLF?	Review of sampling plan regarding sampling media, sampling locations, analytical techniques, and QA/QC issues	Preliminary results
B.2 Which other factors need to be addressed in EPA's evaluation of the Superfund status for the NTLF site?	Review whether NTLF operations will be typical during sampling period; review of non-radiological data (e.g. number of affected residents)	Preliminary results
B.3 Should one be concerned about contamination of radionuclides other than tritium?	Review of non-tritium radioactive contamination at LBNL	In progress
C. Historical Exposures (pre-1998)		
C.1 Which exposures to neutron and gamma radiation resulted from LBNL operations?	Review of historical data on neutron and gamma exposures	Preliminary results
C.2 Which exposures resulted from past releases of tritium?	Review of historical data on tritium emission and environmental concentrations	Preliminary results
C.3 Which exposures resulted from past releases of radionuclides other than tritium?	Review of historical data on emissions and environmental concentrations	In progress
D. Risk Related Questions		
D.1 What is the potential health risk from past exposures?	Comparison of historical doses with doses at other sites	In progress
D.2 What is the potential health risk from current exposures of tritium?	Review of updated health Risk Assessment (expected for 2001)	In progress
D.3 What is the risk in case of accidents, such as wildfire?	Review of LBNL Safety Analysis Document for NTLF	In progress

A Exposures from current operations at LBNL

2.1 A.1 Is the tritium inventory at NTLF adequately determined?

Approach
Review inventory data, its accuracy, and relevance to determine the environmental impacts of NTLF.

Findings to date
LBNL is conducting a tritium inventory using DOE's Nuclear Materials Monitoring System (NMMSS). The purpose of this system is to provide accountability of LBNL's use of tritium. The accuracy of the data, however, is very limited for a variety of reasons:

- All numbers are rounded to 0.1 g of tritium, or 96 Ci
- The reporting threshold is 0.05 g of tritium, or 48 Ci
- LBNL's estimate of tritium inventory at NTLF for has an associated accuracy of less than quantity of historical receipts and shipments with an accuracy of less than 20%

Figure 2 shows the NMMSS inventory data along with reported airborne emissions of tritium from LBNL. To allow visual comparison, the reported releases were plotted at midpoint of the respective year. The airborne releases are two to three orders of magnitude lower than the reported NMMSS inventory. There is no clear correlation between the two numbers. During the 1990s, the reported releases of tritium decreased even though the reported inventory did not change much.

Product shipments may be a better indicator of the activities at NTLF that are associated with airborne releases of tritium. While shipments up until 1991 were typically between 100 and 300 Ci per year, reported product shipments dropped to a level around 10 Ci around the mid-1990s. According to LBNL, the values for 1998 and 1999 were 20 Ci and 10 Ci, respectively. This suggests less tritiation activity during the last few years. However, neither tritium shipments nor the tritium inventory appear to be a good indicator for the likelihood of potential releases.

Conclusions and recommendations
The current inventory of tritium at NTLF is reported to be around 13.000 Ci. The potential error in that estimate, however, is greater than 20% and thus exceeds the reported airborne tritium releases. The limited accuracy of inventory data does not allow a reasonable mass balance, which could be used to verify the releases into the environment. It is desirable to improve the accuracy of tritium inventory. With due consideration to limitations expressed above, the inventory data, in connection with other information such as the number of experiments in a given time period, and the amount of tritium in waste streams, will allow to evaluate whether the type of operation at NTLF can be regarded as typical. It is recommended that the accuracy of tritium inventory data be improved in order to facilitate this determination.

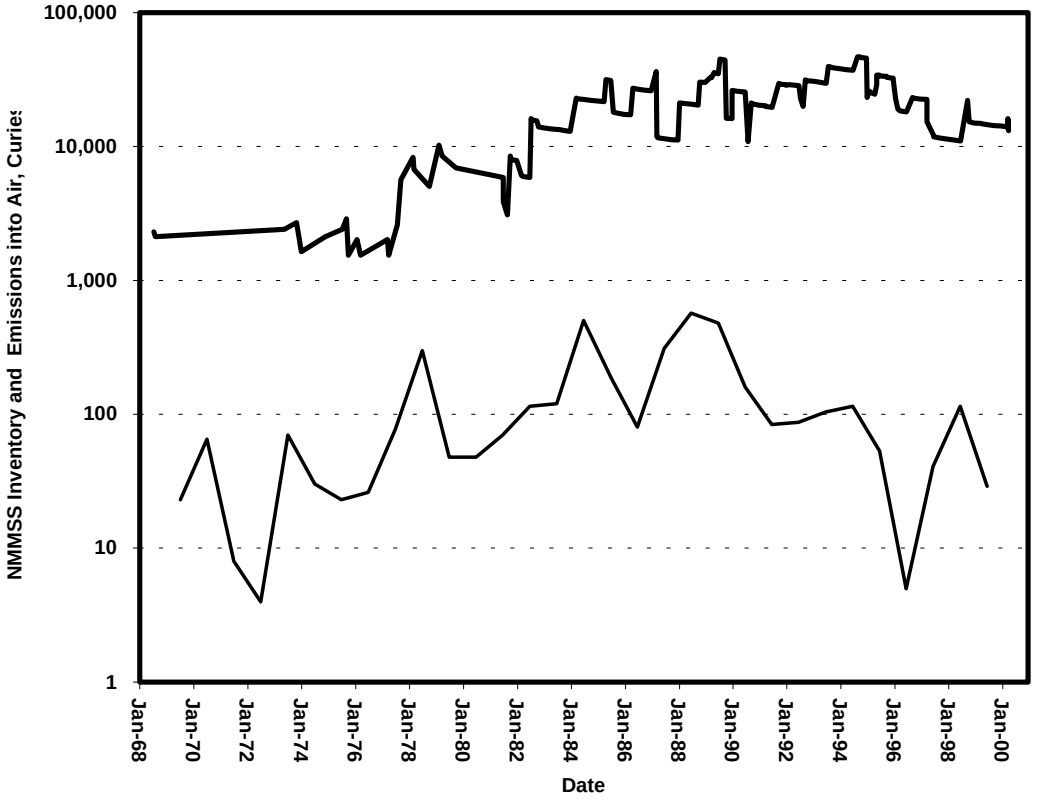

Figure 2. Reported tritium inventory at NTLF (top line) in relation to reported airborne releases of tritium into the air (bottom line)

2.2 A.2 Are the releases of airborne tritium adequately monitored?

Approach
Review data on stack releases; evaluate internal consistency and uncertainties

Findings to date
At NTLF, two independent stack-monitoring systems are in operation. Cumulative weekly releases of HTO and HTO&HT are sampled with a silica gel system on a weekly basis. In addition, a real-time system (Overhoff) provides emission data integrated that is over 100 seconds. The silica sampling distinguished between HTO and HTO&HT since the end of February, 1998, allowing to check the internal consistency of the data since concentrations of HTO should be smaller then those of HTO&HT. LBNL indicates the uncertainty due to sampling and analysis errors to be ~20%, thus the ratio of (HTO&HT)/HTO should be below 1.2. Figure 3 shows the results of the comparison.

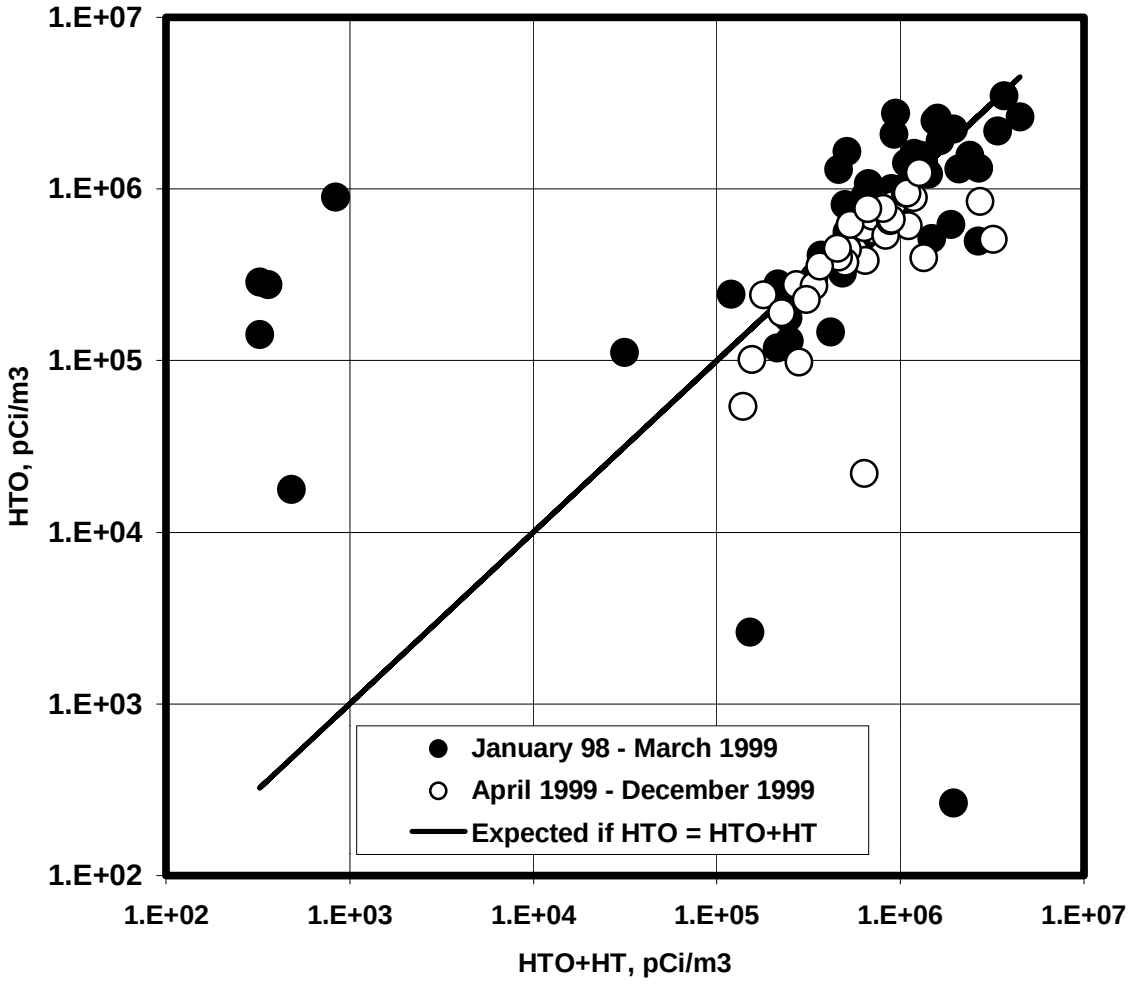

Figure 3. Comparison of concentrations of HTO and HTO&HT determined from weekly silica gel sampling of the NTLF stack

A new oxidizer was installed in March 1998; therefore data before and after this date should be reviewed separately. It is evident that the old oxidizer operating at room temperature was unreliable because 18 out of 57 samples or 32% of the samples had ratios of (HTO&HT)/HTO of greater than 1.2. After April 1999, this number dropped to 1 out of 32 samples, or 3%. When questioned about this, LBNL gave the following explanation: "The oxidizer was initially operated at ambient temperature based on manufacturer's specifications. Some of the air moisture (HTO) was captured (either from condensation or from surface retention) by the oxidizer, which gave a lower tritium concentration value than the HTO column during certain weeks. Since this is a closed system, the captured HTO moisture was eventually being re-evaporated back to the system, which then contributed to a relatively higher reading on subsequent weeks. In fact, we did observe a small amount of moisture captured by the oxidizer cylinder during some of our maintenance activities. Thus, the oxidizer initially behaved like a "delay circuit" which temporally shifted the weekly results. "

This explanation appears to be reasonable. Because the 1998 data for HTO&HT was unreliable, the tritium source term for 1998 in LBNL's NESHAP report was based on silica gel data only with regard to HTO; releases of HT were estimated based on the basis of real-time monitoring system. The data obtained from LBNL was analyzed for the same time periods and plotted in Figure 4. There are two systematic problems with the real-time data for this type of comparison. First, the Overhoff system did not continuously operate for the entire time period due to system malfunctions; the data thus provides only part of the releases. Second, the real-time data is subject to a high instrument background and electrical spikes, which could be falsely interpreted as signals of releases.

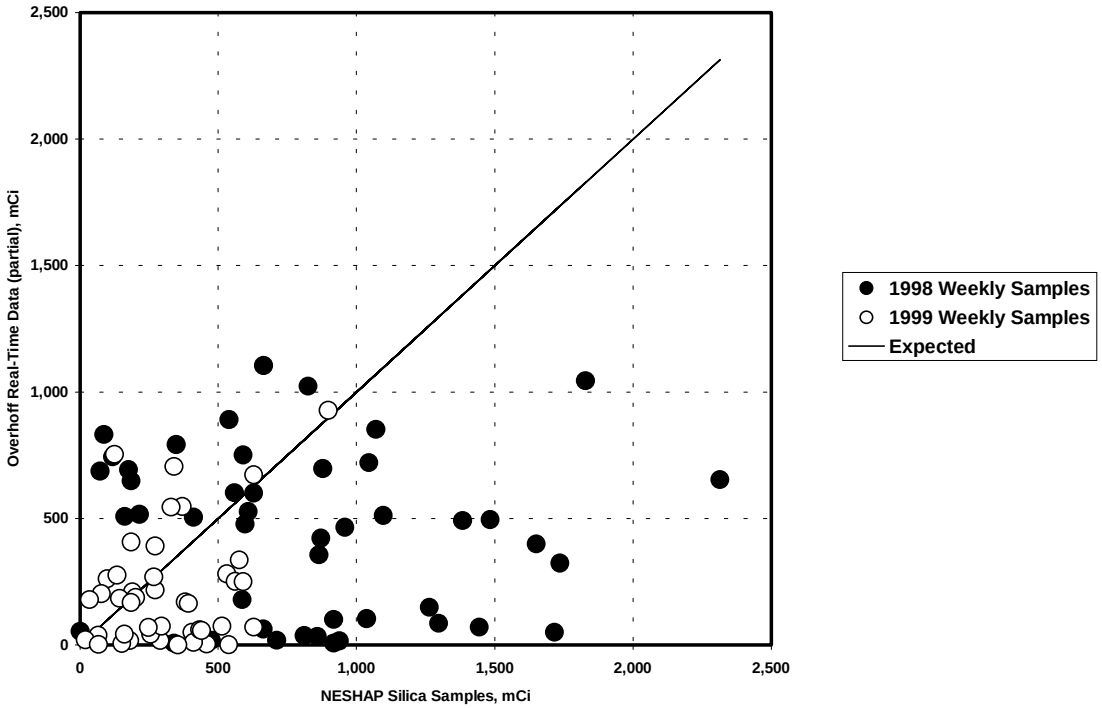

Figure 4. Comparison of weekly releases of HTO determined from weekly silica gel samples and Overhoff real-time measurements at the NTLF stack

The instrument background it is reported by LBNL to be 0.22 Mq/m^3, equivalent to a release of about 4 Ci per week if no background were to be subtracted. Inspection of the real-time data suggests significant fluctuation of the instrument background due to changes in temperature and other factors (see Figure 5). The apparent discrepancy between releases based on silica gel data and integrated releases based on real-time data after nominal instrument background subtraction does not allow an independent verification of silica gel data with real-time data. Even though it may appear that for a number of weeks real-time data suggests larger HTO releases than silica gel data, this finding may well represent an artifact. Fluctuations of the average weekly instrument background could be an explanation of the discrepancies evident in Figure 4. The comparison, however, suggests that real-time data is not a reliable indicator of the quantitative releases from NTLF stacks. Thus, the 1998 releases of HT from NTLF stack, which was estimated on the basis of Overhoff data to be 21.4 Ci is subject to significant uncertainties. The authors were unable to verify this estimate based on the data provide; it is possible that actual releases were smaller or larger than 21.4 Ci. One should bear in mind, however, that for dose assessments, all the tritium sampled, regardless of chemical form, is assumed to be HTO. This assumption is conservative, hence the exact amount of HT released is of minor importance.

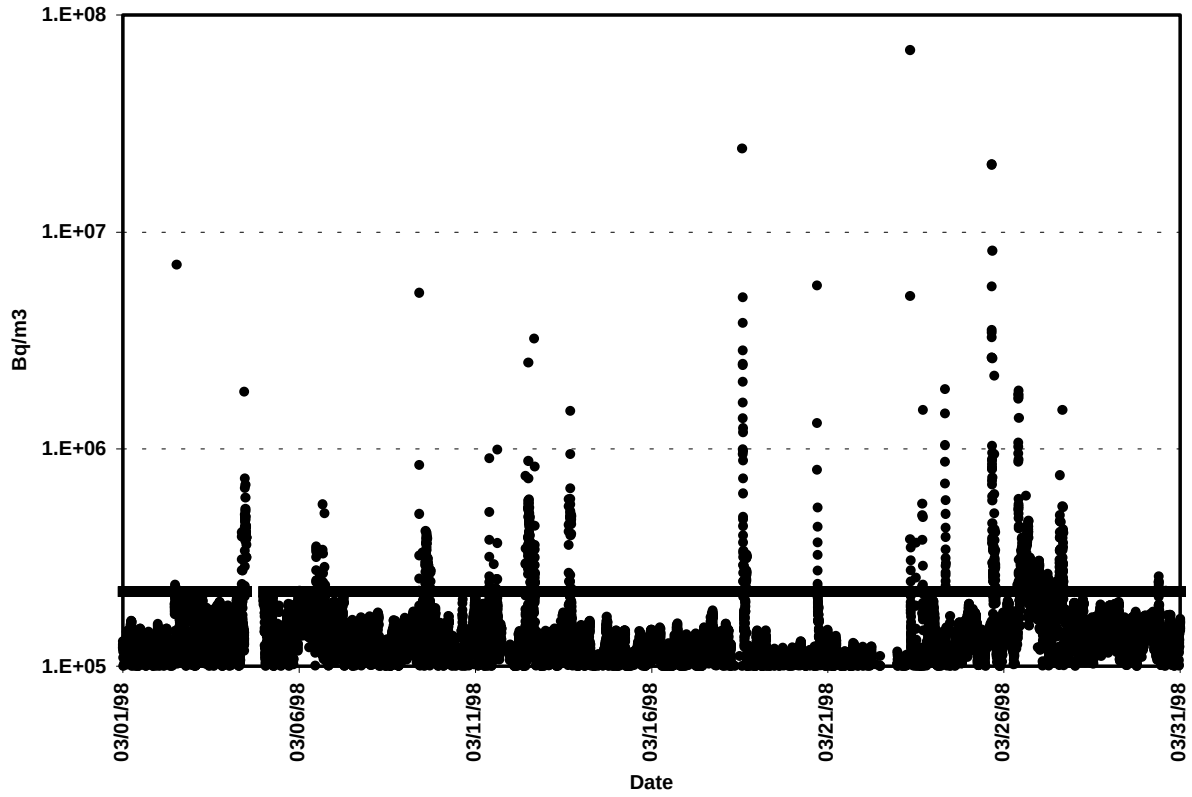

Figure 5. Real-time Overhoff monitoring of HTO releases from NTLF stack, March 1998. The reported instrument background of 220,000 Bq/m^3 is indicated by a

horizontal line. Visual inspection suggests that actual background may have been lower.

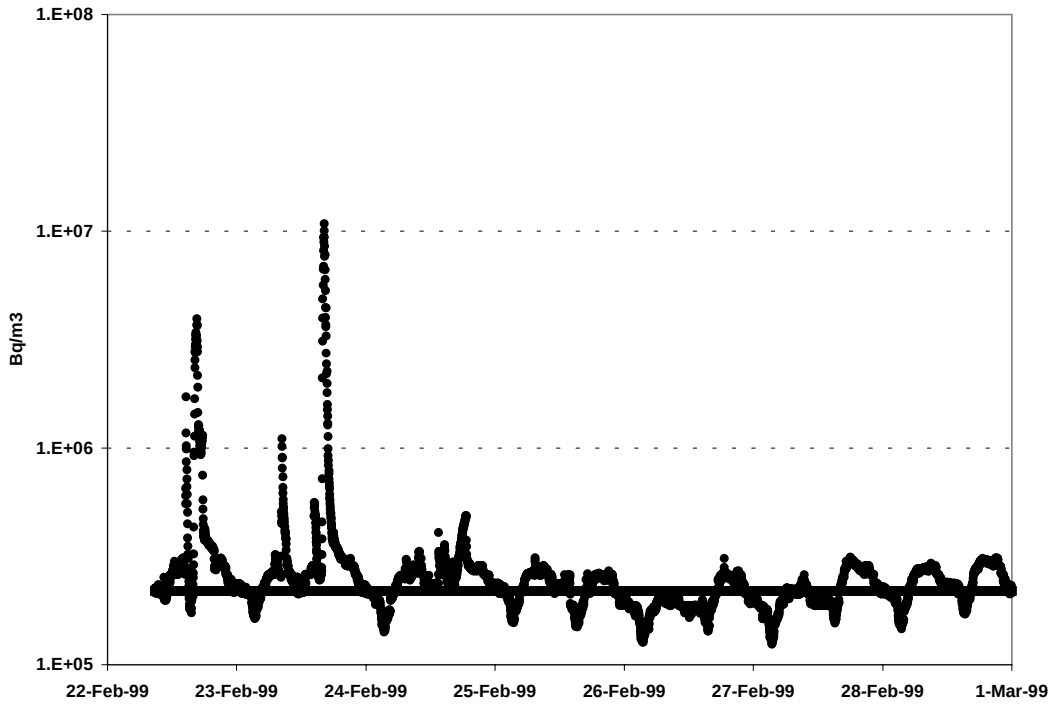

Figure 6. Real-time Overhoff monitoring of HTO releases from NTLF stack, February 22 to March 1, 1999. The reported instrument background of 220,000 Bq/m^3 is indicated by a horizontal line.

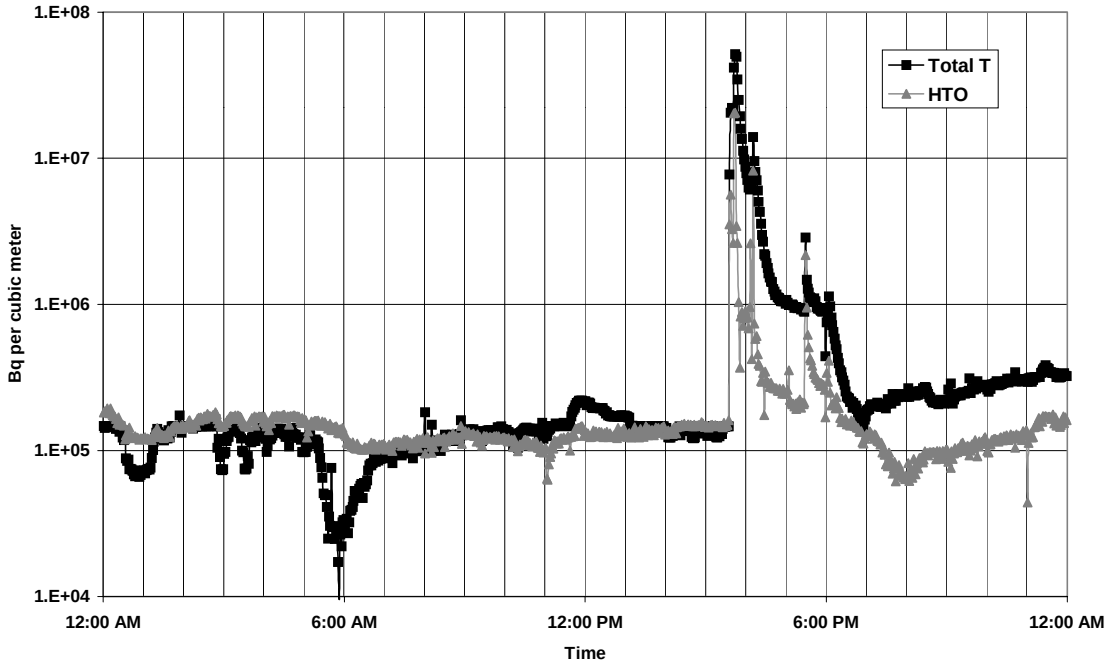

Figure 7. Real-time concentrations of HTO and total tritium in NTLF stack releases on March 25, 1998 (instrument background not subtracted)

Conclusions and recommendations

The review indicates some uncertainties in NTLF stack data for 1998. While the silica gel data for HTO appears to be reliable, the silica gel data for the sum of HTO&HT was unreliable because oxidizer malfunctioning. The real-time data measured with the Overhoff system cannot be used to verify the source term measured with the more sensitive silica gel system. Overhoff data was used to estimate the 1998 release of HT. This estimate is subject to significant uncertainty. However, the relevance of this finding is limited due to low radiotoxicity of HT.

The most significant conclusion is that real-time data shows that tritium is often released from NTLF in short events. On March 25, 1998, a total of 0.29 Ci of HTO was released based on Overhoff data. Of this, 0.2 Ci or 69% was released over a short time period of 1000 seconds (about 17 minutes). Inspection of real-time data suggests that similar patterns are not uncommon for other days as well. A systematic analysis of the time-release function is recommended. Future atmospheric dispersion modeling of NTLF releases should take the short-term nature of the releases properly into account. However, there is no evidence at this time to suggest that offsite exposures resulted in radiation doses exceeding the 10 mrem/yr limit for any individual.

2.3 A.3 Is tritium in air measured at the right locations?

Approach
Compare potentially affected locations with locations actually sampled

Findings to date
During 1999, tritium in air was monitored at six monitoring sites. Three of the sites are on Berkeley Lab property, and three are on the adjacent UC property. With the beginning of January 11, 2000, an additional station (ENV-75 EG) is sampled at 21 meters distance from the stack near the fence in the general direction of LHS. In addition, two additional locations will be sampled once the Tritium Sampling Plan becomes effective, the UC Botanical Garden and the Summit Reservoir, the latter one is selected as background station. The rationale for locating air-sampling stations takes into account the predominant wind direction. In addition, the station at LHS allows verifying calculated doses for the NESHAP compliance location.

One can evaluate the sampling network at LBNL by comparing it to the monitoring systems at other DOE facilities. Table 1 contains the summary data for LBNL as well as for five other facilities for which 1998 tritium emissions were reported to be between 0.054 Ci (Pantex) and 82,700 Ci (Savannah River Site). The number of ambient air monitoring stations for tritium of non-LBNL facilities ranged from 17 to 52. The reported dose to the maximally exposed individual (MEI) which reflects, among other things, the distance to the source of the release, varies greatly between the facilities. Despite the large release at the Savannah River Site, the calculated MEI dose is lower than for LBNL because of the distance between the source and the fence (greater than 10 miles). Based on the data in Table 3, the LBNL site has the smallest number of sampling locations for tritium in ambient air even though the distance from the source of tritium emissions to MEI is nowhere as close as it is at LBNL.

Table 3. Ambient air monitoring for tritium at DOE facilities

Facility	1998 Tritium Release (Ci)[a)]	Number of ambient air stations[b)]	1998 calculated dose to facility MEI from all radionuclides and sources combined
E.O. Lawrence Berkeley National Laboratory	115	6 (+3)[c)]	0.28 mrem
Los Alamos National Laboratoy	818	52	1.1 mrem
Brookhaven National Laboratory	39.5	22	0.21 mrem
Savannah River Site	82,700	17	0.08 mrem
Pantex	0.054	27	0.005 mrem
Lawrence Livermore National Laboratory	110	20	0.055 mrem

a) Based on Site Environmental Reports for the respective facility
b) according to Baumann (2000)
c) see text

Visual inspection of the sites suggests that the sampling network at most facilities is covering all 16 wind direction sectors. It thus appears reasonable to adopt a similar design for for LBNL as well.

An additional reason for increasing the number of monitoring sites at LBNL is the fact that tritium releases occur over comparatively short time periods. Hence, these emissions may be dispersed in directions other than what would be expected from the average distribution of wind directions. This makes the precise selection of appropriate stations complicated. If, say, 1 Ci of tritium would be released during a time period of a few minutes, there is a significant chance that the current network of monitoring stations may not properly detect it. If a person happens to be close to the fence during such an event in downwind direction, he or she could receive a dose, which could be larger than the one calculated with CAP88PC for Lawrence Hall of Science. The probability of this coincidence could be determined with appropriate dispersion model calculations. Up to this point, there is no indication that exposures actually exceeded the 10 mrem/yr dose limit for any individual.

Conclusions and recommendations

The number of sites that are monitored for tritium in ambient air should be increased to cover at least all 16 wind directions. The selection of precise locations should be based on a detailed evaluation of expected tritium concentrations in air using a dispersion model capable to account for the complex terrain and the short-term nature of tritium releases.

2.4 A.4 Is the sampling and analysis of tritium in air at a given location sufficiently accurate?

Approach
Review observed versus expected water collected in silica gel samples
Review results of split sampling program
Review of contract laboratory performance

Findings to date
Silica gel sampling of tritium in ambient air will only return adequate information if all water which is in air is actually captured on the gel. Observations of the sampling system at Los Alamos National Laboratory showed that less water was collected than expected from humidity measurements in air. This problem was especially pronounced in dry summer months. The authors asked LBNL to provide a comparison of the amount of water actually collected in the silica gel samples with the amount expected in the volume of air passing through the gel during the same time period. The expected moisture was estimated using data from the onsite meteorological station. The result is shown in Figure 8 using ENV-69 station as an example. There is a reasonable correlation between the two data sets. The ratio (observed/expected) for a monthly sample was in a range of 0.77 to 1.28, for the annual average the range was 0.97 to 1.08.

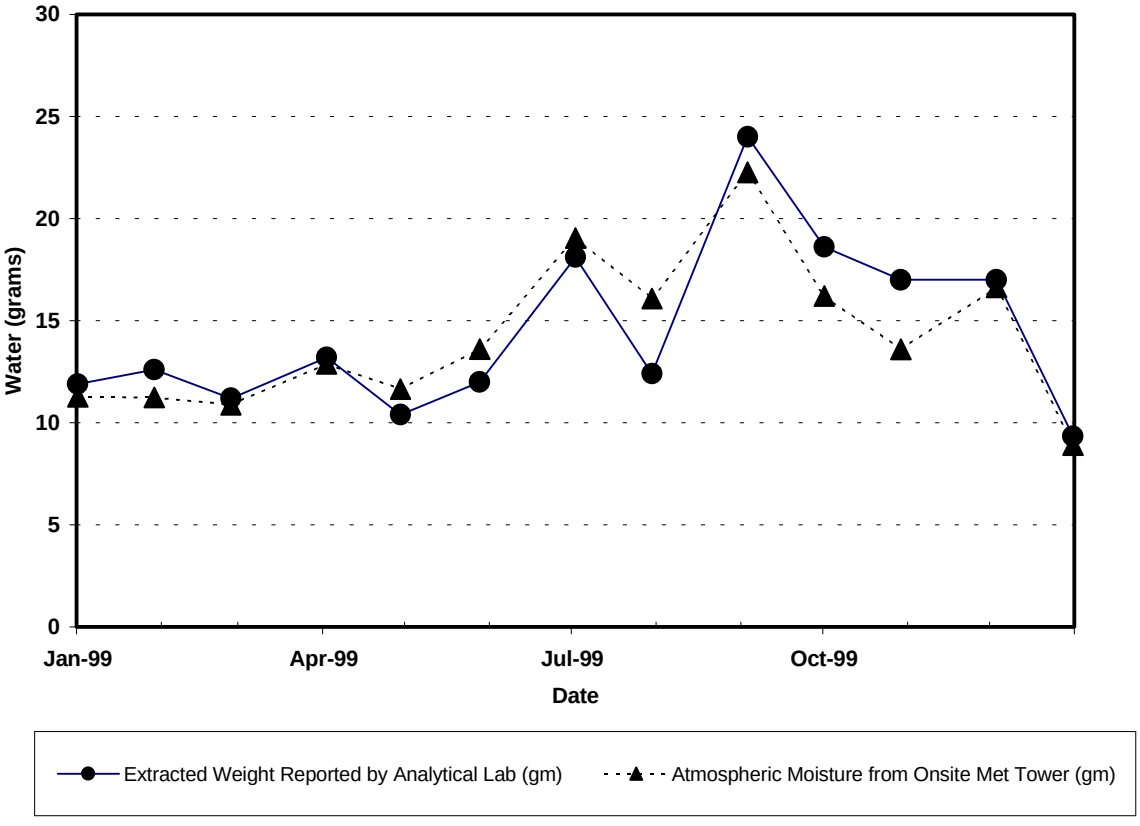

Figure 8. Comparison of observed and expected water collected at ENV-69

Figure 8 indicates that in some months, the amount of water extracted from the silica gel exceeded the amount expected from onsite met tower data. This may well be due to the fact that silica gel has an initial water load prior to environmental sampling that is driven out in the distillation process in the laboratory. This additional amount of water can be in the order of several grams. It is therefore suggested that the weight of water collected in silica gel sampling be determined by the difference in silica column weight before and after the sampling period. The weight difference rather than the amount of water extracted should be compared with the amount of water expected from the onsite meteorological data.

Since November of 1997, silica gel samples are split and analyzed both by LBNL and with the EPA laboratory. The results for two stations are shown in Figure 9 and 10. As expected, the correlation is better for station LHS due to higher concentrations of tritium. In the annual average, the values reported by EPA are 20% higher for the station ENV-13D and 3% for ENV-LHS, the uncertainty is larger for individual weekly samples.

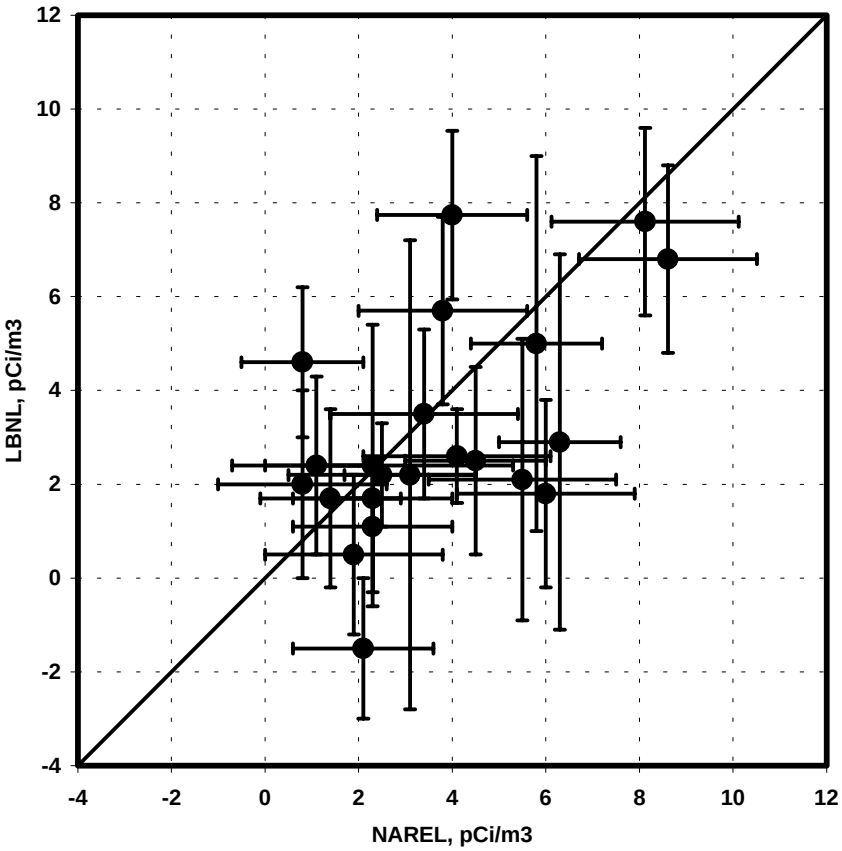

Figure 9. Comparison of tritium split samples EPA's of NAREL and LBNL analytical laboratories for station ENV-13D

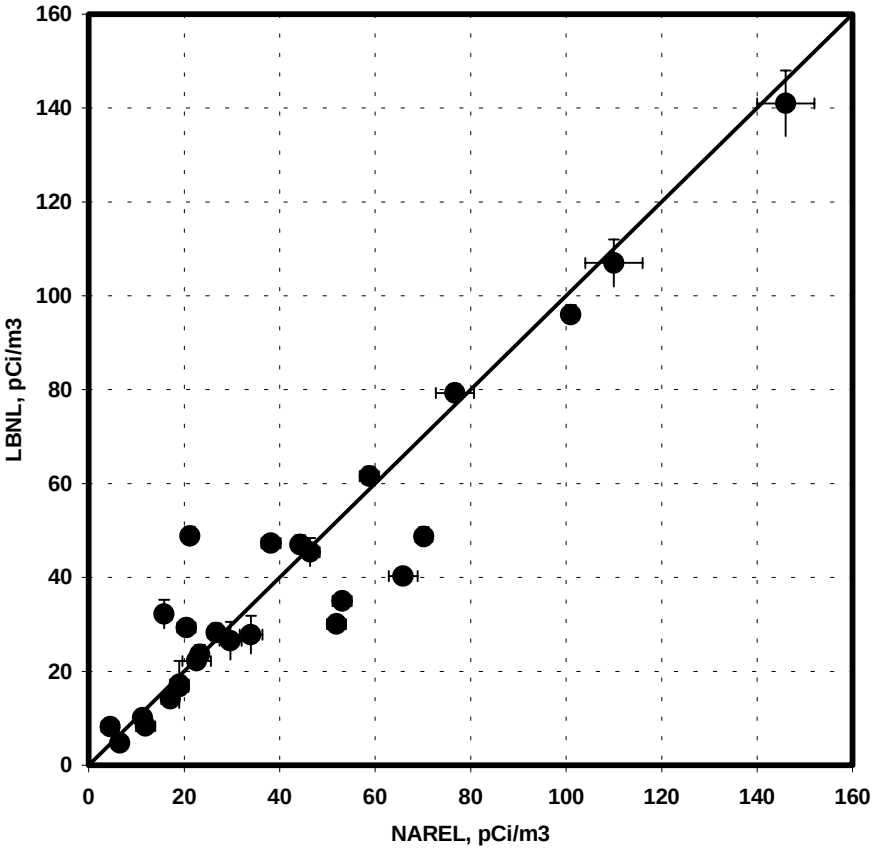

Figure 10. Comparison of tritium split samples of EPA's NAREL and LBNL analytical laboratories for station ENV-LHS

Spot checks of the Tritium Monitoring Analytical Laboratory Data for 1998 which was provided to IFEU by LBNL indicates that the sampling and analysis data are properly documented and that the calculations based on this data are verifiable.

Conclusions and recommendations

Based on the data reviewed thus far, the analytical data for HTO in ambient air samples is verifiable and is subject to reasonable uncertainties. With regard to analysis of samples collected at the Lawrence Hall of Science, the annual average concentration of tritium appears to be subject to a combined uncertainty of less than 20%. It is suggested that information regarding the uncertainty of analytical data be incorporated in the site environmental reports. It is suggested that the amount of water collected in silica gel be determined from the sampler weight difference.

3 B Legacy contamination from past operations / Superfund issues

3.1 B.1 Is LBNL's Draft Tritium Sampling and Analysis Plan sufficient to determine the extent and nature of legacy contamination at NTLF?

Approach
Review of sampling plan regarding sampling media, sampling locations, analytical techniques, and QA/QC issues.

Findings to date
The Draft Tritium Sampling and Analysis Plan currently addresses the following environmental media:

- ambient air
- soil
- sediment and surface water
- vegetation

The EPA hazard ranking for the NTLF site indicates that **ambient air** data will be most decisive for the evaluation. It is therefore necessary to properly address the issues in section A.2 to A.4. It is advisable to expand the network of ambient air sampling stations to cover all 16 wind direction sectors (of 22.5° each). This will ensure that LBNL site is sampled with a network of comparable density to that at other DOE facilities. It is not sufficient to focus on the predominant wind direction sectors because tritium releases from NTLF often occur over very short time periods during which direction of the wind can differ greatly. The placement of the stations should be optimized pending the results of the proposed modeling with a complex terrain and conservative release patterns for NTLF.

The proposed sampling sites for **soil** cover all areas that can reasonably expected to have been contaminated. However, given the fact that large tritium releases may have occurred during wind directions with a low frequency, emphasis should be placed on the representative sampling of the area around each site. Rather than the proposed single sample from each site, ten cores should be taken according to the core method in DOE's Environmental Measurements Laboratory Procedures Manual HASL-300, section 2.3.4.1 (EML, 1997). An additional requirement of this procedure is to select undisturbed sites if possible. The sampling plan is further deficient in providing a rationale to limit sampling to one depth increment of 15 to 30 cm. In order to provide a representative results for all soil depths which are potentially contaminated, it would be advisable to collect the following depth increments: 0 to 15 cm, 15 to 30 cm, and 30 to 60 cm, and 60 cm to 150 cm.

The proposed program of **sediment and surface water** and **vegetation** sampling is well designed. No changes appear necessary.

The program should be expanded by analysis of **groundwater** samples as well. Groundwater sampling, although not essential for EPA hazard ranking of the site, would respond to community concerns regarding the potential risk from the site.

The NTLF site may not be the only one affected by past operations. Integrated concentrations of tritium in ambient air at Building 3 (Calvin) on the UC campus are comparable to those measured at NTLF, which could indicate a similar level of environmental contamination (see section C.2). A preliminary soil sampling effort would be advisable for the surrounding of Building 3.

Conclusions and recommendations

The Draft Tritium Sampling and Analysis Plan sampling and analysis program should be supplemented as follows:

- expansion of ambient air monitoring to cover all 16 wind direction sectors (of 22.5° each)
- use of HASL-300 core method for soil sampling, samples to be analyzed for additional depth increments
- sampling of groundwater in coordination with the State of California Water Resources Board
- preliminary sampling efforts around Building 3 (Calvin)

B.2 Which other factors need to be addressed in EPA's evaluation of the Superfund status for the NTLF site?

Approach
Review whether NTLF operations will be typical during sampling period; review of non-radiological data (e.g. number of affected residents)

Findings to date
EPA will assess the score of the LBNL site using the Hazard Ranking System (HRS) scoring process. There are valid concerns in the community that operations at NTLF during the sampling time may not be representative of typical operations. This issue was addressed in section A.1. It was determined that the tritium inventory does not serve as a good indicator of laboratory activity due to the large uncertainty inherent in the data. It is therefore recommended that an array of information be used in the determining whether NTLF is operations are representative including the shipment of products and number of tritiations performed.

EPA concluded in its preliminary assessment that the Lawrence Hall of Science (LHS) is not regarded as a school. While this is a correct finding, one should not neglect the fact that LHS is visited by more than 300,000 visitors per year, a third of which are students. If LHS would qualify as a school, the number of students would be entered in the HRS scoring system. In order to determine whether this assumption is significant in HRS scoring, it is proposed that EPA provide two separate sets of scoring calculations, one of which assuming LHS as a school, accounting for the full-time equivalent visitor population plus resident staff. Alternatively, the number of repeat visitors should be determined.

Conclusions and recommendations
- Inclusion of a section describing NTLF operations during sampling time when reporting the results
- EPA should provide information as to how the hazard ranking score would change if Lawrence Hall of Science would be regarded as a school, accounting for student population.

4 C Historical exposures (pre-1998)

4.1 C.1 Which exposures to neutron and gamma radiation resulted from LBNL operations?

Approach
Review of historical data on neutron and gamma exposures

Findings to date
At early times neutron exposures of employees (as well as possible offsite exposures) were significant. Professor E. O. Lawrence himself requested that the Physiology Dept. at U. C. Berkeley look into the possible harmful effects of neutrons after finding the at the building which housed one of his cyclotrons had become activated by neutrons. The accelerators at Lawrence's lab were used primarily for research in high-energy physics, but also (and later at LBNL) for radiation biology, medical research, atomic physics, isotope production and research, and very high intensity photon sources. Operating accelerators produce a variety of radiation fields outside of the biological shielding which is intended to protect personnel from radiation exposures. These are primarily neutrons, gamma rays, muons, and other radiations of which neutrons have the highest intensity, and are the most damaging from a health risk point of view.

The Laboratory's environmental monitoring reports from 1960 to 1976were reviewed. During this period the Bevatron accelerated protons and other light ions to energies, which often exceeded 6.4 GeV, the maximum endpoint energy for protons. Neutron production was incidental to the acceleration of light ions, and because of their lack of electrical charge, they penetrated the thick concrete shielding to produce exposures in persons both on-site and off-site. The spectrum of these neutrons was best described by the function 1/E, where E is the neutron energy. One can derive a neutron field through thick shielding for a proton beam at 6.4 GeV such that the neutron spectrum would extend from "thermal" energies (average of 2.5 E–8 MeV) to 6.4 E3 MeV.

From the Olympus Gate monitoring station the Bevatron looked like a point source producing a neutron field described by 1/E up to 6.4E3 MeV. Superimposed on this 1/E spectrum was the contribution from "sky shine". This was the result of high-energy neutrons escaping the shielding in a roughly vertical direction, and interacting with molecules of air, resulting in their being scattered back to the ground at substantially reduced energies (=10 MeV). The sky shine neutron spectrum declined as roughly 1/r from the Bevatron, whereas the direct neutron spectrum declined as 1/rE2, where r is the distance from the Bevatron.

Early environmental reports indicate that maximum doses for the Olympus Gate station were 810 mrem from neutron and gamma radiation in 1959 and of 650 mrem in 1960 (Figure 11). These exposures were in excess of the dose limit of 500 mrem per year set in AEC Manual Chapter 0524, dated February 1, 1958. It appears that LBNL was subject to AEC Manual Chapter 0524 regulations since the laboratory was an AEC contractor. The Manual chapter 0524-02 paragraph 2 states that existing facilities can apply for a conversion period not to exceed five years if a request is made by an appropriate AEC official. IFEU has asked the

LBNL to supply a written copy of the request if it was made at the time. Such a document has not been received to date. Thus, the issue remains unresolved whether LBNL doses in unrestricted areas exceeded then-prevailing limits using dosimetric methods in use at the time.

A recent LBNL report (Thomas et al., 2000) provides a reassessment of neutron doses using different conversion factors of neutron spectra to dose than used in the past by LBNL. That report makes a credible case that for reducing the earlier reported doses by a factor of at least two (2), as shown in Figure 11.

Radiation doses calculated for any of a variety of recipient conditions could vary a great deal. No attempt was made to calculate doses to persons beyond the site boundary, but rather to keep the boundary doses within acceptable limits. The best estimate of the impact of neutrons on the environment is a description of the neutron spectrum as a function of energy (the differential energy spectrum). Issues relating to exposure of persons to that neutron field may have a profound effect on the description of radiation dose and consequent health risk from that exposure. For example, the dose itself can be calculated for persons facing the source (AP), away from the source (PA), laterally from the side of the body (LAT), rotating with respect to the source (ROT), or exposed to an isotropic source (ISO), ICRP (1997) and ICRU (1998). Additionally, shielding may be provided by a housing structure, for instance. These issues combined with the uncertainty associated with residency times can force a dramatic impact on dose estimates in the public sector. Hence, the decision about minimizing the dose at the site boundary.

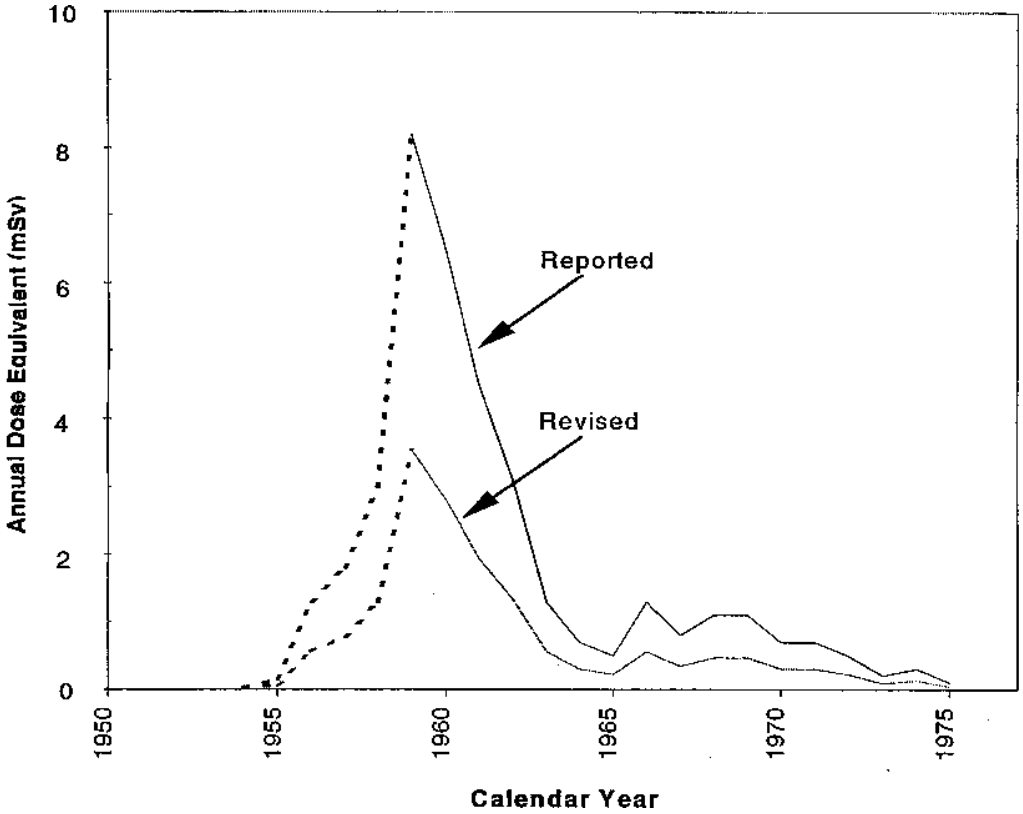

Figure 11. Comparison of reported dose equivalents for 1959-1975 with revised dose equivalents (Thomas et al., 2000)

Conclusions and recommendations

Neutron and gamma doses at various locations at the LBNL site boundary were substantially larger than today. Peak exposures may have exceeded then-prevailing limit of 500 mrem/yr using the historical conversion factors. Using current conversion factors for neutron doses, cumulative doses at the Olympus Gate station were in the order of a few rem. It is recommended to estimate doses to nearby residents while taking uncertainties and contribution all sources and pathways into account. The fact that historical doses of comparable magnitude were assessed in detail at other DOE sites may serve as justification for this effort.

4.2 C.2 Which exposures resulted from past releases of tritium?

Approach
Review of historical data on tritium emission and environmental concentrations

Findings to date
Measurements of HTO in ambient air for three sampling locations are shown in Figure 12 for the time period 1969 to 1999 along with reported tritium emissions. Starting in 1995, emission data includes HT as well as HTO while before that, only HTO emissions were reported. Three observations can be made on this basis of this data. First, the ratio of reported tritium concentration in ambient air to reported releases is consistently larger for the time period 1969 to 1986 (>10 pCi/m^3 per Ci/yr) compared to the 1987 to 1999 (<10 pCi/m^3 per Ci/yr). Second, the concentrations at Building 3 (Calvin) were equal to or larger than levels measured at Lawrence Hall of Science (LHS). The peak concentrations in 1985 of 3,000 pCi/m^3 was more than a factor of 100 greater than the concentrations measured at LHS in 1999 and exceeded the current NESHAP compliance standard of 1,500 pCi/m^3. Third, reported concentrations at LHS and Olympus Gate were often equal to those reported for LHS while one would expect lower concentrations due to atmospheric dispersion.

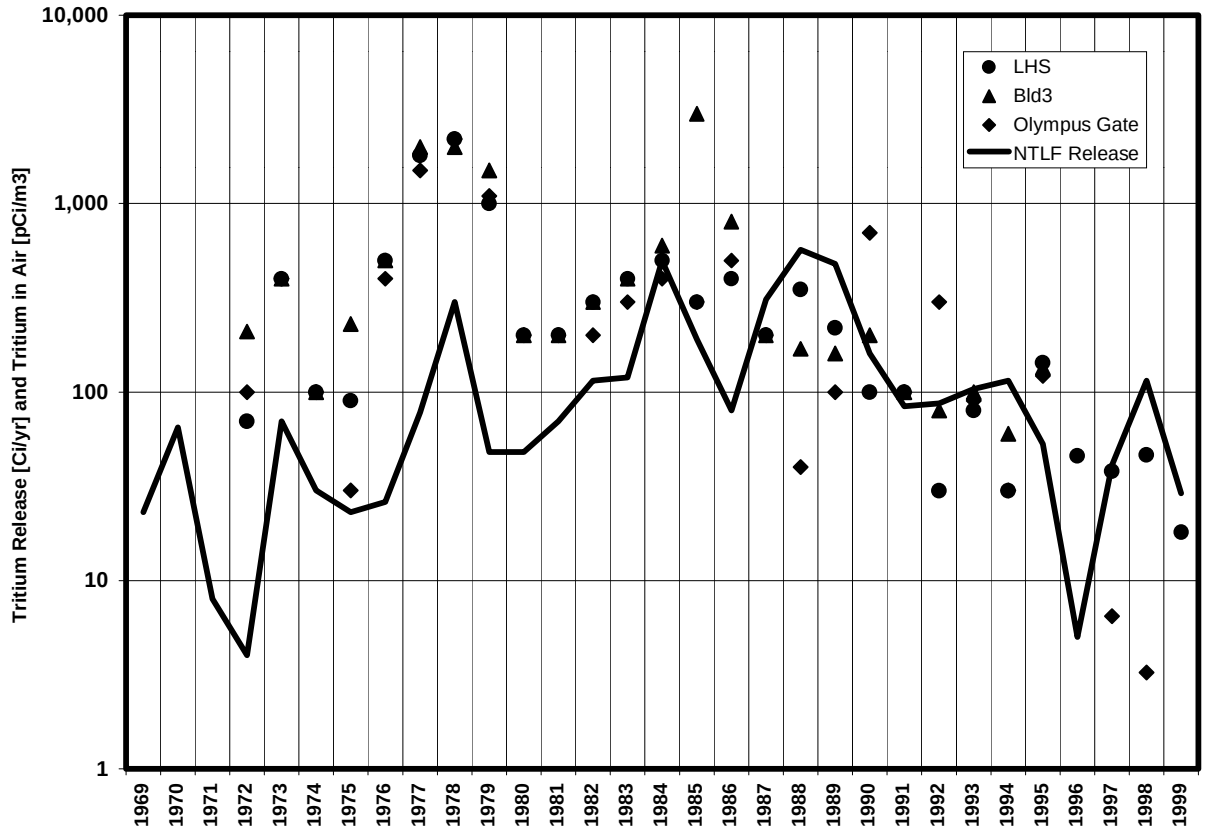

Figure 12. Reported annual tritium releases from NTLF, 1969 to 1999 and annual concentrations of HTO in air at environmental monitoring stations

In Figure 13, reported tritium emissions are plotted against the concentrations reported for Lawrence Hall of Science (LHS). For years with releases around 100 Ci/year, reported annual average air concentrations differ by a factor of ~100, indicating a significant variability in ambient air concentration for a given annual emission. This may be caused by difference in year-to-year meteorology, analytical errors, the impact of short-term releases and other factors.

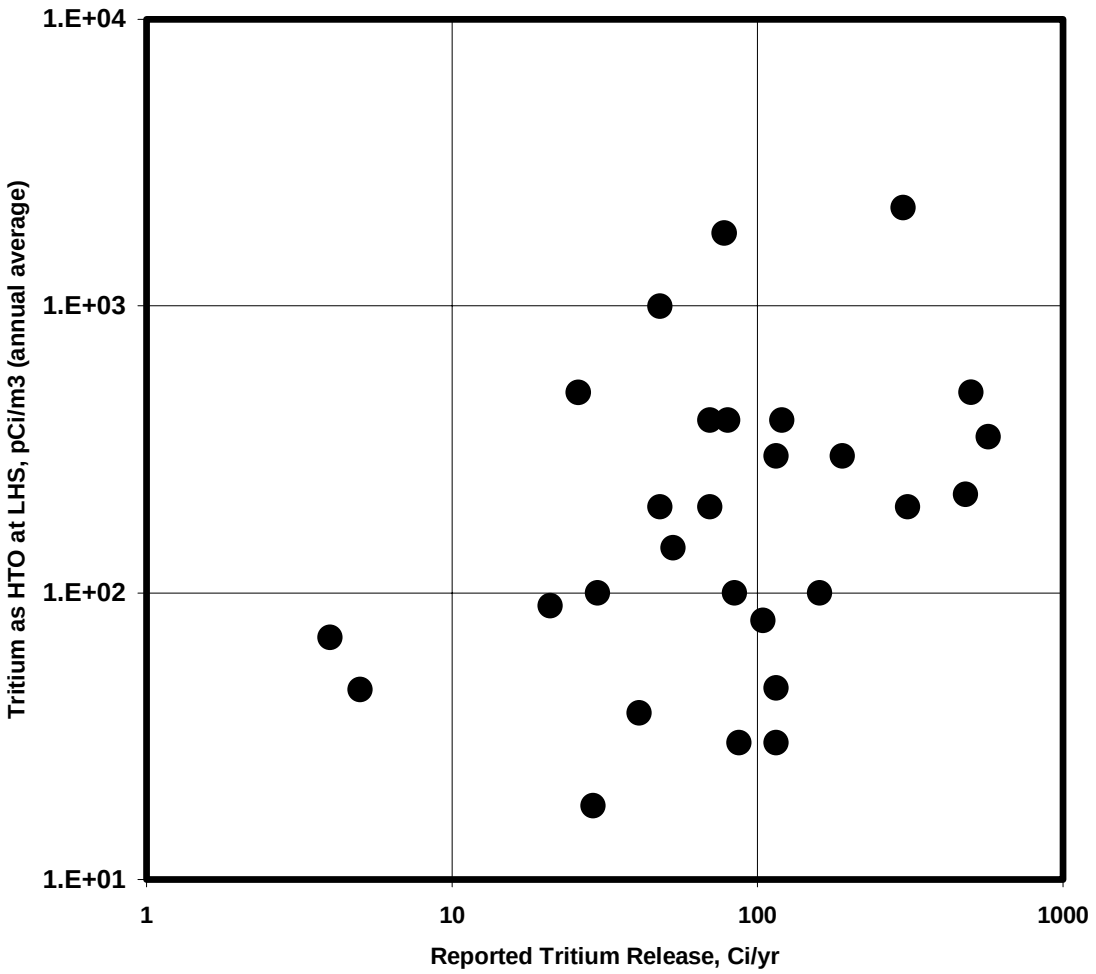

Figure 13. Reported annual tritium releases from NTLF, 1972 to 1999 and annual concentrations of HTO in Air at Lawrence Hall of Science (LHS)

Conclusions and recommendations

Taking the data at face value and assuming that sampled locations were representative, it does not appear that current dose limits are exceeded. However, the accuracy should be evaluated in light of the fact that pre-1995 measurements are considered unreliable. This evaluation should include:

- Review of uncertainties of data regarding tritium emissions and ambient monitoring

- Compare reported concentrations with those expected from dispersion modeling using complex terrain
- Review the conditions around Building 3 on UC campus

5 References

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Appendix A

SCOPE OF SERVICES

1. Work Plan

1.1. Contractor shall interview LBNL technicians and scientists, subcontractors, regulators, City officials, and representatives of the community to generate a list of concerns for evaluation. Identify the data quality objectives (DQO) from the list of concerns.

1.2. Contractor shall review sufficient representative data on present and past emissions and environmental monitoring by LBNL of soils, air, subsurface and ground waters, and plants (including raw data) in order to arrive at conclusions about both the quality of the data collected and the analyses of that data.

1.3. Contractor shall review and comment on the LBNL/U.S. Department of Energy (DOE) tritium sampling work plans for air, water, soils, plant, and other media.

1.4. Contractor shall review the adequacy and accuracy of LBNL's past monitoring, analyses, and modeling.

1.5. Contractor shall review and comment on the appropriateness of models used for calculations of doses and risks, including confirmatory sampling calculations, as necessary.

1.6. Contractor shall provide recommendations on future data collection and further investigations at LBNL, including dose reconstruction, if necessary.

1.7. Contractor shall review and provide a written report on the revised health risk assessment documents that are expected to be produced by LBNL/DOE at the end of the process.

1.8. Deliverables:

1.8.1. Report #1: Contractor shall produce a **Preliminary Technical Report** on the evaluation of past and present emissions and environmental data. The report shall include a review of the identified DQO and a review of the sampling plans.

1.8.2. Report #2: Contractor shall produce a **Draft Final Scoping Report**, which shall include a review of the appropriateness of models used for the calculations of doses and risks, with recommendations for further investigations, if warranted. The report shall be submitted to the City, which shall be responsible for distribution of the report to stakeholders, as well as for compiling all written comments for transmittal to Contractor.

1.8.3. Evaluation of Comments: Contractor shall review the written comments on the draft reports (both those submitted directly to Contractor and those compiled by the City of Berkeley) and revise the reports as necessary. The result of this task shall be the **Final Scoping Report**. Contractor shall submit this report to the City for distribution to the stakeholders.

1.8.4. Report #3: Contractor shall produce a report on the results of the sampling plan, including a qualitative review of the LBNL's revised health risk assessment documents. The report shall be presented to the City, which shall distribute it to the stakeholders. Written comments received by Contractor or collected by the City will be reviewed and updates made as appropriate.

1.8.5 Presentation. A presentation of the **Final Scoping Report and Report #3** (Deliverables 1.8.3. and 1.8.4.) shall be made before the Berkeley City Council by Contractor and others.